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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re-application of:

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MIWA, Yasutaka DOI

Application No.: 10/589,501

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For: MODIFIER FOR RESIN AND RESIN COMPOSITION USING THE SAME AND  
FORMED ARTICLE

DECLARATION UNDER 37 CFR § 1.132

COMMISSIONER FOR PATENTS

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U.S.A.

Sir:

I, Tsuneki WAKITA, hereby declare and state that:

1. I am a citizen of Japan, 20-1, Miyuki-cho, Otake-shi, Hiroshima, Japan
2. I am fully familiar with the subject matter of the subject application as well as the references relied upon by the Examiner in the prosecution of this application.
3. I obtained a Master's degree in Applied Chemistry from Nagoya University Graduate School in 2002.
4. I am currently employed by Mitsubishi Rayon CO., LTD., and began working



for Mitsubishi Rayon CO., LTD. on 2002. Since 2002, I have been engaged in the research of Impact Modifiers for thermoplastic resins and thermosetting resins in the Resins & Plastics Development Center.

5. I conducted the following experiments.

### **I. Object**

The following experiment demonstrates that the polymer disclosed in Rauch et. al. does not meet the requirement of that of the present invention.

### **II. Samples**

#### **(Reference)**

##### **Preparation of Modifier for Resin (IM-5)**

In a 5 liter flask, 119 parts of pure water, 5 parts of butyl acrylate and 0.125 parts of allyl methacrylate were charged and heated to 80 °C while stirring at 250 rpm in a nitrogen atmosphere.

A previously prepared solution of 0.15 parts of potassium persulfate and 6.0 parts of pure water were charged at a time and the first stage soap-free emulsion polymerization was conducted while maintaining for 60 minutes. Then, a mixed solution of 65 parts of butyl acrylate, 1.625 parts of allyl methacrylate, 0.68 parts of sodium di 2-ethylhexylsulfosuccinate (manufactured by Kao Corporation under the trade name of PELEX OT-P) and 38.7 parts of pure water was added dropwise over 180 minutes and the second stage emulsion polymerization was conducted while maintaining for one hour to obtain an acrylic rubber polymer latex (R-5). To the resulting latex (R-5), a mixed solution of 29.4 parts of methyl methacrylate, 0.6 parts of ethyl acrylate, 0.51 parts of sodium di 2-ethylhexyl sulfosuccinate and 17.9 parts of pure water was added dropwise over 100 minutes and, after maintaining for one hour, the emulsion polymerization was terminated to obtain a graft copolymer (G-5) latex. The resulting latex had an average particle size of 250 nm. The amount of cullet during the



polymerization was 0.16% The resulting graft copolymer latex was sprayed by a spray dryer with a pressure nozzle type and the resulting microdroplets were dried at a hot-air inlet temperature of 180 °C to obtain a modifier for resin (IM-5).

The resulting powder had an average particle size of 36  $\mu\text{m}$ . The powder particles had a disintegration property of 1 %. The rubber moiety had a glass transition point of -25 °C. and the graft moiety had a glass transition point of 83 °C.

### III. Tests

The measurement result of the average particle size of the latex used in the preparation of the resulting modifier for resin (IM-5), the average particle size of the modifier for resin before and after irradiation with ultrasonic wave, the content of particles having an particle size of 10  $\mu\text{m}$  or less and the glass transition point are shown in Table 1 below.

100 parts by mass of a curable resin was mixed with the modifier for resin to obtain sheet-like formed test pieces. Using the resulting test pieces, Izod impact strength and dispersibility of the modifier were evaluated.

#### (Impact Strength)

A sheet-like test piece was formed, cut and then evaluated in accordance with ASTM D256. (Thickness: 1/4 inch, Unit: J/m)

#### (Dispersibility)

Dispersion state (coagulation state) of a modifier for resin on the surface of a sheet-like test piece was visually evaluated.

### IV. Test Results

Table 1



A resin for Impact Strength	Average particle size of latex [nm]	Average particle size of powder				Glass transition [°C]	
		Before Irradiation with ultrasonic wave		After Irradiation with ultrasonic wave [40W x300sec]		Rubber moiety Shell moiety	
		Content of particles having a size of 10µm or less [%]	Average particle size [µm]	Content of particles having a size of 10µm or less [%]	Average particle size [µm]		
IM-2	600	3	38	57	7	-25	88
IM-5	250	1	36	1	36	-25	83

Note: IM-2 (Example 2 of the present invention) is shown for comparison.

Table 2: Evaluation of the dispersibility

The sheet-like test piece was applied to liquid nitrogen and freeze-fractured.

The fracture cross-section was observed by SEM and a dispersibility of a resin for impact strength was evaluated.

	A resin for Impact Strength	Izod Impact strength test [J/m]	Dispersibility
Example 2	IM-2	22	A
Reference	IM-5	15	B

A: Particles having a size of 10 µm or more is not observed.

B: Particles having a size of 50 µm or more is not observed, but particles having a size of 10 µm or more is observed.

Note: IM-2 (Example 2 of the present invention) is shown for comparison.

## V. Conclusion

The average particle size of 250 nm in the latex (IM-5) which corresponds to the sample disclosed in Rauch et. al. was both inferior in Izod impact strength test and dispersibility to the average particle size of 600 nm in a latex (Example 2, A modifier IM-2) of the present invention.

Accordingly, Rauch et. al. is not capable of achieving the present invention.

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6. I understand fully the content of this declaration.

7. The undersigned declares that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

8. Further declaration saith not.

Tsuneki Wakita

Tsuneki WAKITA

October 7, 2011

Date